Interface Issues in Solid and Gel Polymer Electrolytes for Sodium Batteries

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Abstract

Lithium-ion batteries, commonly used in many portable devices, are popular due to their high energy output and versatile applications. However, there is a need to develop an alternative due to the high cost of lithium and ethical concerns about lithium mining. Sodium-ion batteries are a promising substitute due to sodium's similar chemical properties to lithium, abundance, low cost, and environmentally friendly extraction process. However, scientific advancements in sodium-ion battery technologies have been impeded for a number of reasons: mainly the interface instability between the electrolyte and sodium anode. Interface instability can occur when chemical reactions at the interface cause the battery to short circuit and when sodium dendrites form in the voids at the interface with the sodium metal and electrolyte. In an attempt to address these dendrite issues, researchers have turned to solid state electrolytes. Additionally, liquid electrolytes, commonly used in lithium-ion batteries, cannot be used in sodium batteries due to safety issues; thus gel and solid state electrolytes (SSEs) are especially important in the context of sodium batteries. This paper intends to determine what specific problems, with an emphasis on interface stability, are seen in solid polymer electrolytes (SPEs) and gel polymer electrolytes (GPEs) while addressing strategies to broaden the applicability of sodium-ion batteries.

1. Introduction

Since lithium ion batteries (LIBs) were introduced to the consumer market in 1991, they have become an essential and widely used energy source across many industries, from portable devices to electric vehicles. While they have many advantages, such as high energy density, long cycle life, and lightweightness, resources of lithium and cobalt, necessary for $LiCoO_2$ batteries, are dwindling at a rapid rate. For example, to generate 95 million electric vehicles, it would take an estimated 50% of all natural lithium reserves (Yang et al.). On average, 67 million vehicles are sold each year worldwide, so if all of these cars were electric, the lithium supply would run out in 2-3 years (Statista). Lithium extraction also causes environmental damage in the form of air, water, and soil pollution. The toxic chemicals and immense water, 500,000 gallons of water per ton of lithium extracted, used during lithium mining can harm the miners and locals through water shortages and chemical poisoning (Kaunda). The Chilean Atacama region, rich in copper, gold, silver, and lithium, is one of the driest desert regions of the world, yet miners are still continuing to extract extreme amounts of groundwater. 0.00005-0.001 g of lithium needs an estimated 1 liter of water and a lithium ion battery needs around 4.4 g of lithium (Agusdinata et al.). Therefore, each lithium battery would require around 4400-88000 liters of water to manufacture. In addition, as mining activity increases, the ecosystem deteriorates, because lithium mining includes a process in which the lithium is washed with sodium carbonate in polyvinyl chloride-lined ponds. Cracks in the lining result in the leakage of chemicals into the environment. Moreover, for every 1 Watt-hour of LIB storage capacity, 110 g CO_2 equivalent of greenhouse gas emissions are released (Agusdinata et al.).

On the other hand, sodium is an environmentally friendly material and a naturally abundant metal with the ratio of lithium reserves to sodium reserves being 1:1000 (Fig. 1)

(Vignarooban et al.). Sodium also has similar chemical properties to lithium since they are both alkali metals, making the transition from lithium batteries to sodium batteries favorable from a scientific discovery standpoint. Some sodium-ion batteries (SIBs) are on the market like NaeS and $NaeNiCl_2$ (ZEBRA e Zero Emission Battery Research Activities) batteries, but these require high temperatures of 300° C in order to operate, which hinders their real world applications (Vignarooban et al.).

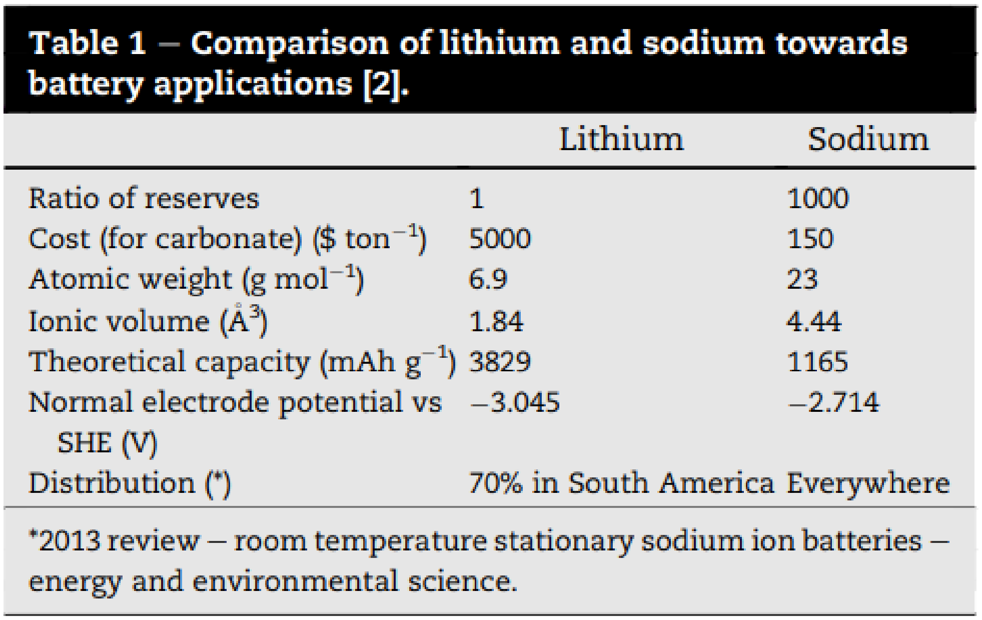

Table 1 – Comparison of lithium and sodium towards battery applications [2].

	Lithium	Sodium
Ratio of reserves	1	1000
Cost (for carbonate) ($ ton^{-1})	5000	150
Atomic weight (g mol^{-1})	6.9	23
Ionic volume ($Å^3$)	1.84	4.44
Theoretical capacity (mAh g^{-1})	3829	1165
Normal electrode potential vs SHE (V)	−3.045	−2.714
Distribution (*)	70% in South America	Everywhere

*2013 review – room temperature stationary sodium ion batteries – energy and environmental science.

Fig 1: Table of sodium versus lithium properties taken from (Vignarooban et al.)

Developing proper electrolyte and electrode materials are crucial to the further development of SIBs (Vignarooban et al.). Sodium batteries are not compatible with conventional liquid electrolytes (LEs) because LEs are prone to leakage and flammability, making them a severe safety hazard (Yang et al.). Therefore, efforts have been redirected toward solid state electrolytes (SSEs) (Zhao et al.). Polymer electrolytes (PEs), a type of SSE, have good flexibility and processability, which means they have the ability to be applicable in many fields. The two types of PEs are solid polymer electrolytes (SPEs) and gel polymer electrolytes (GPEs) (Wang et al.). SPEs have good flexibility and low flammability but ultimately suffer from low ionic conductivity at RT (Zhao et al.), which is considered one of the most important properties of an electrolyte. GPEs tend to have higher ionic conductivities than SPEs but have poor mechanical strength (Wang et al.).

There are a few features that an ideal electrolyte should have (Fig. 2B). For performance, the electrolyte should have high ionic conductivity, high stability, good mechanical strength, a stable interface, and dendrite resistance. For applicability, it should be environmentally friendly, cost-effective, and easy to manufacture. Since many of the issues impeding further development of PEs are related to the interface (interface stability, dendrite formation, and low ionic conductivity), the further development of sustainable batteries can be achieved through rigorous research of interface issues (Lu et al.). In this review, we will discuss past and current developments on stable interfaces for SIBs using SPEs and GPEs. We will discuss how interface stability, dendrite formation, and ionic conductivity affect the practicality of SIBs.

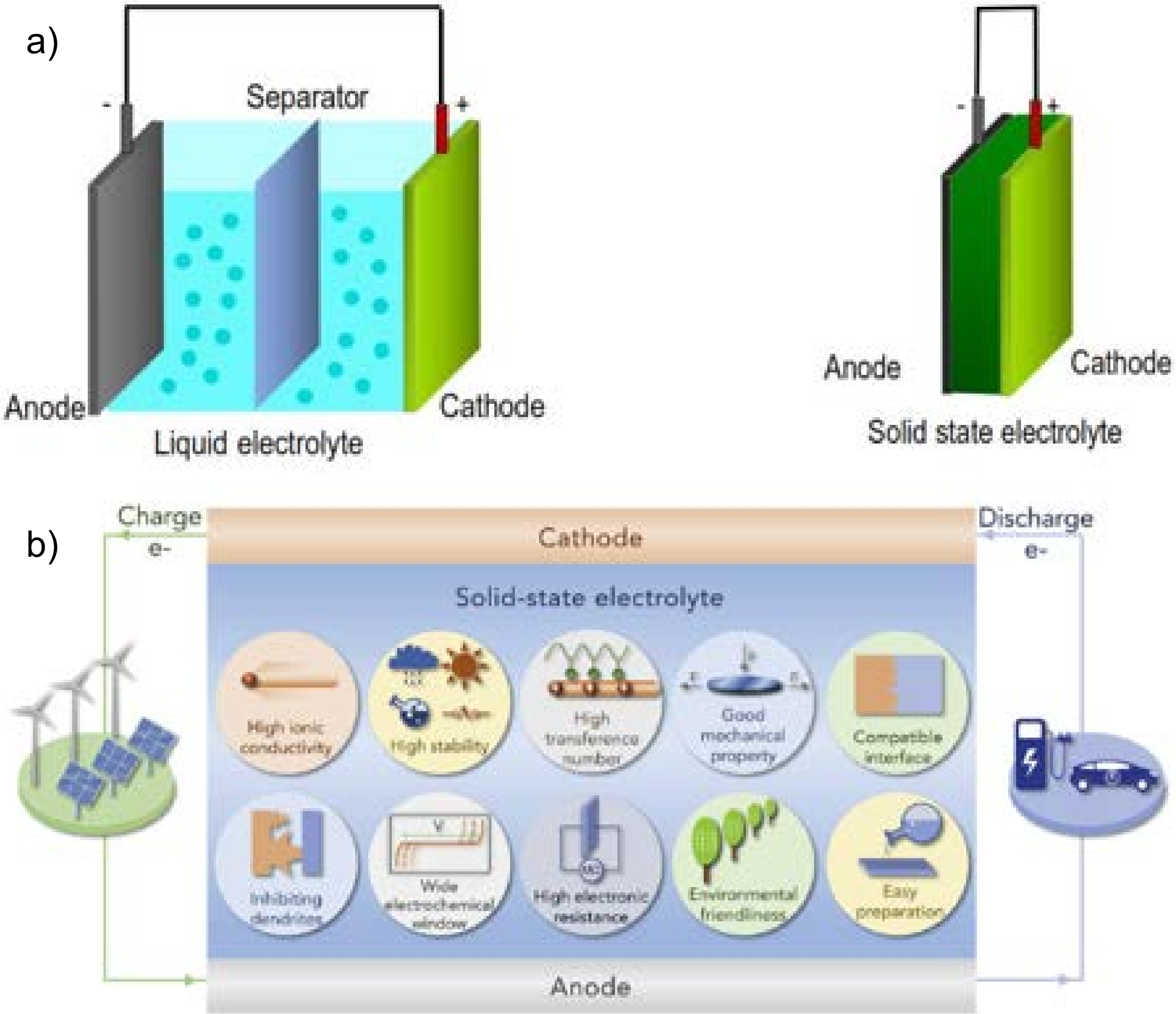

Fig 2: a) Diagram depicting basic parts of a battery taken from (George). b) Diagram depicting ideal characteristics of a solid state electrolyte taken from (Lu et al.).

2. The Interface Issues in Sodium Ion Batteries

One of the biggest issues in the development of sodium ion batteries (SIBs) is interface stability (Hou et al.; Lu et al.). The materials of the anode and electrolyte will often react, and these reactions can cause electrolyte decomposition, create the solid electrolyte interphase (SEI) layer, and even go as far as to completely alter the properties of the electrolyte (Lou et al.; Sawicki and Shaw). Normally, SEIs in metal batteries have high mechanical stability, will protect the electrolyte from decomposition, and separate the anode and electrolyte effectively. However, in SIBs, the SEI layer has poor mechanical stability, leading the battery to malfunction. During the stripping/deposition process, the volume expansion of the Na^+ anode causes non-uniform SEI layer formation, which leads to cracks in the SEI layer. These cracks enable dendrite growth and lead to short circuiting (Fig. 3). They also rip small pieces off of the Na anode and expose a fresh SEI layer that further reacts with the electrolyte (Bao et al.; Lee et al.; Lou et al.).

While sodium and lithium are similar since they are both alkali metals, they still have different physical and electrochemical properties that may make the transition from LIBs to SIBs difficult. Na^+ is more reactive because of its larger atomic size, so rapid transfer of electrons from the anode to electrolyte causes the growth of an unstable SEI, resulting in sodium dendrite growth, electrolyte consumption, and interface resistance (Lee et al.).

Moreover, it was discovered that poor mechanical and electrochemical stability could lead to the SEI layer disappearing. The Na anode and SEI layer's poor electrochemical and mechanical stability produce gas in the cell, putting the entire electrolyte at risk of complete decomposition. Several investigations into the gas evolution have been done, and it has been discovered that the gas evolution has detrimental effects on the battery (Lee et al.).

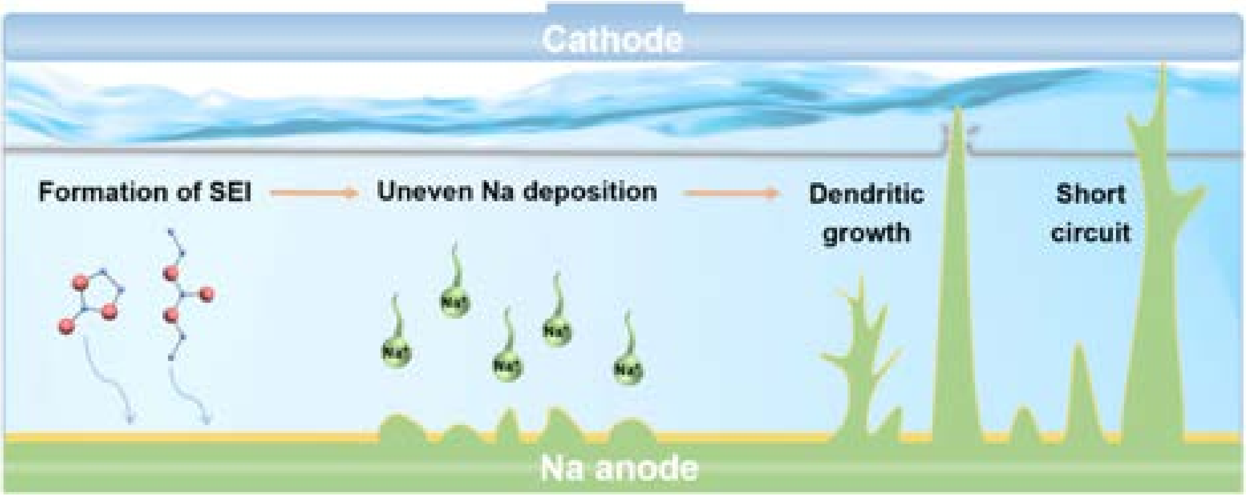

Fig 3: Diagram of sodium dendrite formation from Li et al. (2022)

Dendrites are currently one of the most difficult issues that impede the commercialization of SIBs, because they not only lower the battery efficiency, but they also present a major safety issue. For example, dendrites can lead to short circuiting, which can cause the battery to catch fire or explode. The deformities in the sodium metal form an uneven electric field that causes the Na deposition to crack the SEI layer. The exposed metal causes another SEI layer to form. The continued deposition under the Na metal consumes the metal and creates dendrites (Li et al.).

Dendrites have been an issue since LIBs conventionally contained liquid electrolytes. SSEs were initially anticipated to be able to overcome the dendrite issues due to their solid quality, but they have been observed to facilitate dendrite formation. This quality has been reported in all SSEs, including polymers, sulfides, and oxides, but it is especially a concern in SPEs due to polymer's low mechanical strength (Lou et al.). While lithium dendrites are too strong to be suppressed by an SEI layer, sodium dendrites are soft enough that a mechanically strong SEI layer could effectively suppress them, which many strategies like adding ceramic fillers crosslinking have successfully achieved (Lee et al.).

3. Solid Polymer Electrolytes and Interface Issues

Polymer electrolytes (PEs), first investigated by Wright et al. (1973), have recently started to be implemented for the development of SIBs, and since then, many different polymer hosts have been explored (Lou et al.). One of the first and most investigated polymer hosts, poly ethylene oxide (PEO), was developed from the research of Wright et al. (1981) and is popular for its flexible segments, raw material accessibility, chemical stability, and high ionic conductivity at higher temperatures. However, it has been reported to have low ionic conductivity at room temperature (RT) around 10^{-5} S cm^{-1}, poor mechanical qualities, and low oxidation potential (see Fig. 4) (Lou et al.; Yang et al.).

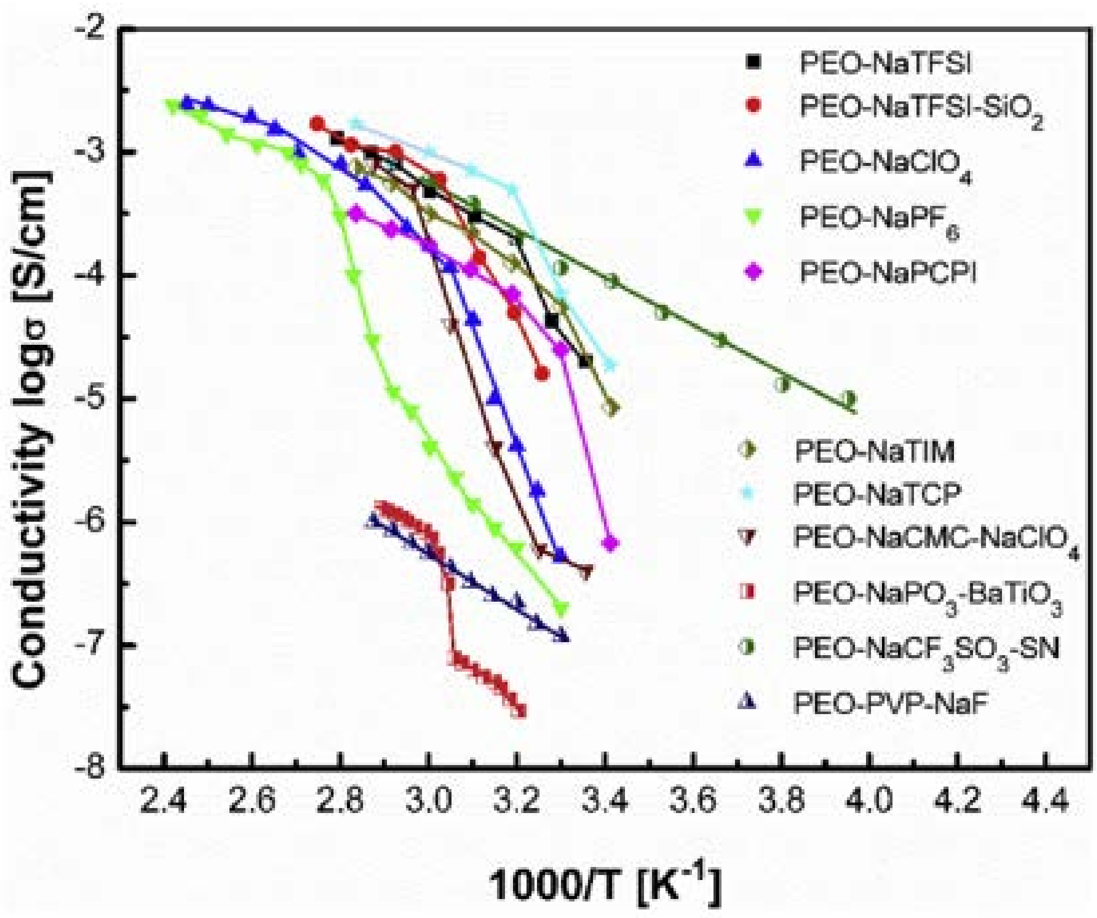

Fig 4: Graph of conductivity of PEO-based SPEs taken from Wang et al. (2019)

Some other common polymer hosts are poly(vinylidene fluoride) (PVDF), poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP), PMMA, PVA, and poly(acrylonitrile) (PAN). Each polymer host has unique advantages and disadvantages (see. Fig. 5). PAN is a popular polymer host for its higher ionic conductivity (~~7.13 x 10^{-4} S cm^{-1}) (Zhao et al.). However, it exhibits low mechanical strength and flexibility. PAN has a rigid structure resulting in low flexibility and low ion transport. PVDF has good thermal stability and wide electrochemical windows but also suffers from low ion transport to optimize their interface properties to allow for higher ion transport. PMMA and PVA are also promising but are nowhere close to replacing PEO (Lou et al.). PVA electrolytes have also been investigated for their higher ionic conductivity (1.36 x 10^{-4} S cm^{-1} at 40 degrees C) (Zhao et al.). By optimizing the ratios, NaBr-PVA electrolytes have shown a higher ionic conductivity of 1.36 x 10^{-4} S cm^{-1} at 40 C. Overall, SPEs are a promising option for SSBs due to their flexibility, but their low ionic conductivity at RT ultimately limits their real world application (Zhao et al.).

Characteristics of Common Polymer Hosts for SPEs

Polymer Host	Reported Ionic Conductivity (S cm^{-1})	Positive Properties	Negative Properties
PEO	10^{-5} (RT) (Yang et al.)	- Widespread commercial availability - Chemical stability (Zheng et al.)	- Low mechanical strength (Zheng et al.)
PVDF	1.1×10^{-4} (RT) (Qiao et al.)	- Good electrochemical and chemical stability - Good thermal stability (Zhao et al.)	- Low ion transport (Zhao et al.)
PVDF-HFP	$>10^{-4}$ (Ling et al.)	- Good mechanical properties - Suppress dendrites (Ling et al.)	N/A
PMMA	1.9×10^{-3} (30 °C) (Qiao et al.)	- Higher ionic conductivity (Zheng et al.)	- Poor mechanical strength (Zheng et al.)

Fig 5: Table with reported ionic conductivity in specified conditions and characteristics of common polymer hosts.

SPEs often suffer from low ionic conductivity due to an insufficient Na^+-ion transference number (>0.5) (Yang et al.). Therefore, several strategies like co-polymerization, blending, and crosslinking have been implemented and seen to improve the ionic conductivity at RT. However, while these electrolytes exhibit high ionic conductivity, they tend to lack other qualities, including mechanical strength or electrochemical stability (Yang et al.). Moreover, many attempts to optimize battery performance have been made by optimizing combinations and concentrations of Na salts, blending it with other polymers, or adding inorganic nanoparticles (Yang et al.). The main Na salts that have been employed with PEO electrolytes are sodium chlorate, sodium perchlorate, sodium polyphosphate, sodium bromide, and sodium iodide. The concentration of Na salts are crucial in optimizing ion-conducting abilities of PEO-based electrolytes at RT (Yang et al.). To further optimize ionic conductivity, (Yang et al.) has tried blending PEO with other polymers. For example, a poly(vinylpyrrolidone) (PVP) based battery has been reported with an ionic conductivity of up to 1/9x10^{-6} S cm^{-1} below RT. Another PVP blended polymer electrolyte system had a reported ionic conductivity of 1.07 x 10^{-5} S cm^{-1} at RT (Yang et al.). Using a polymer blend of PEO and PVDF, a flexible solid electrolyte membrane has been created with an ionic conductivity of 2.34 x 10^{-6} S cm^{-1} at RT. The flexibility and amorphous nature allows for enhanced Na-ion conductivity and electrochemical stability (Augustin et al.). Another SPE including PVDF-HFP has good mechanical properties, a high ionic conductivity above 10^{-4} S cm^{-1}, and is able to effectively suppress dendrites (Ling et al.).

Many strategies have been proposed to correct some of the interface issues seen in SPEs. Since the SEI layer directly depends on the material of the electrolyte, careful thought goes into the composition of the electrolyte to better control variables such as ionic conductivity, strength, and stability. (Lee et al.) has suggested a strategy to put additives into the electrolyte to increase flexibility, encourage high ionic conductivity, and encourage a dense and uniform formation of the SEI (Lee et al.; Sawicki and Shaw). An ideal SEI layer should also be mechanically stable to allow for effective electron transport but no side reactions (Lu et al.).

Nanostructured hosts like Cu foam, carbon matrix, and Mxene have been suggested to help prevent volume expansion of the anode during the stripping/deposition process.Volume expansion can lead to the cracking of the SEI layer and dendrite formation (Lu et al.). Several strategies for preparing an artificial SEI layer have been proposed including chemical coating, physical deposition, and free standing films(Shi et al.). Each strategy has its advantages as well as disadvantages. Inorganic artificial SEI layers generally have high ionic conductivity and high mechanical strength but are brittle. Organic artificial SEI layers are flexible, creating uniform contact with the anode, but the mechanical strength is inferior. Hybrid organic-inorganic SEIs have flexibility and mechanical strength but have limited control over distribution of organic-inorganic components (Shi et al.). Another strategy has been proposed to use the reaction temperature, time, and concentration to control the chemical composition, thickness, and structure of a stable artificial SEI layer. Some of these artificial SEIs have reported higher dendrite suppression but lack other properties like mechanical strength (Yang et al.). Therefore, while an artificial SEI layer could help the suppression of sodium dendrites, they can not fix all the issues with Na anodes. Multiple strategies must be employed if SIBs are to be practical (Shi et al.). So far, developing stable SEI formation has been a challenge that needs to be overcome to further the advancement of SIBs (Matios et al.).

(Liu et al.) proposed a strategy to prepare SPEs with a spontaneous two-step chemical reaction to avoid electrochemical reactions at the interface by transferring a residual free solvent into a chemically inert compound. They were able to achieve a high initial Coulombic efficiency of 93.8% along with good capacity retention and long-term cycling performance (Liu et al.).

To combat the interface reactions between Na-metal anodes and PEO based SPEs, a strategy to replace the anode with a Na/C metal anode has been proposed. It is suspected to allow for more homogenous plating and stripping due to the carbon fiber's high surface area, which could also help reduce dendrite formation (Zhao et al.). Also, an SPE with a blend of PEO and sodium bis(fluorosulfonyl)imide {$Na[N(SO_2F)_2]$, NaFSI} had a reported ionic conductivity of 4.1 x 10^{-4} S cm^{-1} and good electrochemical stability. Notably, the SEI layer formed within the first few cycles and was very stable and effectively suppressed dendrite formation (Qiao et al.).

4. Gel Polymer Electrolytes and Interface Issues

SPEs often do not possess the high ionic conductivity at RT for a high performance battery, so GPEs were developed through the use of small molecular plasticizers to gelatinize various polymer electrolytes. In figure 5, the most common polymer hosts are listed along with their defining physical properties. GPEs are promising due to their nonflammability, wide electrochemical stability windows, high ionic conductivity compared to SPEs (10^{-4} S cm^{-1} vs 10^{-6} ~ 10^{-8} S cm^{-1}), and good thermal stabilities (Yang et al.). They also have the unique ability to transport ions quickly similar to LEs, and the cohesive characteristics similar to SSEs (Aruchamy et al.).

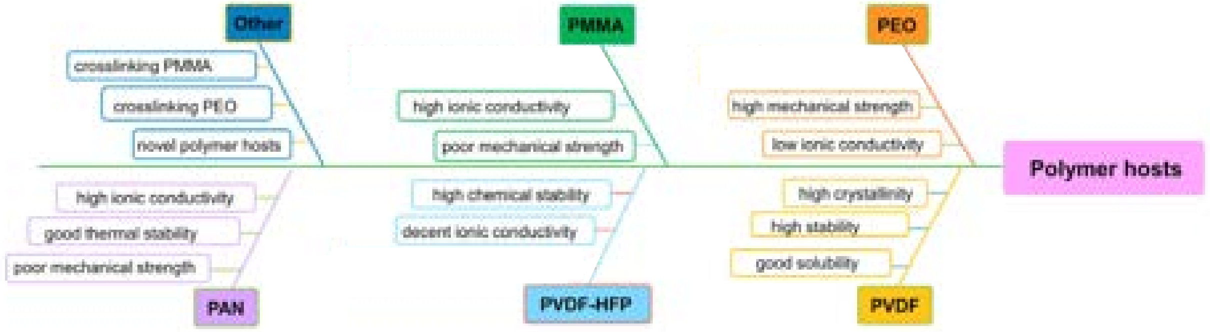

Fig 6: Chart depicting characteristics of common polymer hosts. Modified from Zheng et al. (2022).

Polyethylene oxide (PEO), a common polymer host for GPEs, is popular for its widespread commercial availability (Zheng et al.). Many methods have been employed to optimize the properties of PEO-based GPEs. Scientists using an optimized ratio of a solvent mixture to plasticize the GPE have achieved an ionic conductivity of 9.5 x 10^{-3} S cm^{-1} (Zheng et al.). However, this caused low mechanical strength in the electrolyte. Scientists have used a method called cross-linking to achieve higher mechanical strength, which has a reported ionic conductivity of 2.0 x 10^{-4} S cm^{-1} at 20 C° (Zheng et al.). Another cross-linked GPE with PEO and poly(ethyleneimine) has obtained an ionic conductivity of 1.7 x 10-4 S cm^{-1} at 30 C° (Zheng et al.). Other strategies include using ionic liquids as plasticizers instead of organic and inorganic fillers as polymer hosts have achieved similar promising results. Poly(vinylidene fluoride) (PVDF) has good electrochemical and chemical stability. Poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP) is one of the most popular polymer hosts found in GPEs and has good chemical stability. Polymethyl methacrylate (PMMA) has a higher ionic conductivity but has poor mechanical strength. Blending it with other polymers and using cross-linking copolymerization has been used to improve its mechanical and electrochemical stability (Zheng et al.). Poly(acrylonitrile) (PAN) GPEs have high ionic conductivity of up to 2.35 x 10^{-3} S cm^{-1} at 30°C and good thermal stability (Zheng et al.).

GPEs suffer from many of the same interface issues as SPEs. An ideal GPE should have no side reactions with the anode, a dense and stable SEI layer, and high resistance to dendrites. Often, dendrites can form during the plating/stripping process, and in extreme cases can cause the battery to explode. The SEI layer can protect the battery from side reactions but also increases interface resistance, hindering the battery’s performance (Lee et al.).

Wen et al. (2021) has developed a new gel electrolyte using photopolymerization that has achieved an incredibly high ionic conductivity of 1.2 S cm^{-1} compared to other gel electrolytes with an average ionic conductivity of 10^{-4} S cm^{-1}. The electrolyte exhibits high cycling capacity, no leakage or short circuiting, and no flammability. Their discovery provides a new possible strategy to prepare electrolytes for safe, viable SIBs (Wen et al.). A system created by (Gao et al.) with a Na anode and PMMA crosslinked gel-polymer electrolyte has shown good cycling performance, good ionic conductivity (2.1 x 10^{-2} S cm^{-1}), and stability. It is also promising for large scale energy storage due to its low cost (Gao et al.). By adding graphene oxide, scientists have developed a gel electrolyte that effectively suppresses dendrite growth, is stable, and has achieved an ionic conductivity of 2.3 x 10^{-3} S cm^{-1} (Luo et al.). A poly(vinylidene fluoride-hexafluoropropylene) GPE has uniform SEI formation, dendrite suppression, and no side reactions (Lei et al.). Another crosslinked GPE system has good thermal stability, a high ionic conductivity of 8.2 x 10^{-4} S cm^{-1}, and improved interfacial stability (Niu et al.). Adding hydroxyapatite in PVDF-HDF allows for improved interface stability, reduced leakage, and good ionic conductivity (Niu et al.).

Ionic Conductivities for Common Polymer Hosts

Polymer Host	Reported Ionic Conductivity (S cm^{-1})
PEO	9.5 x 10^{-3}
PEO and poly(ethyleneimine)	1.7 x 10^{-4} (30°C)
PVDF	2 × 10^{-3}
PVDF-HFP	6.0 × 10^{-4} (RT)
PMMA	6.2 x 10^{-3} (RT)
PAN	2.35 x 10^{-3} (30°C)

Fig. 7: Table of reported ionic conductivities of common gel polymer hosts taken from Zheng et al. (2022).

5. Discussion

Sodium ion batteries (SIBs) would be applicable in many fields, such as portable handheld devices and everyday batteries. Recently, the research on and development of SIBs has increased dramatically such that SIBs can compete with LIBs in aspects of battery performance (Sawicki and Shaw). LIBs used in cell phones, computers, and electric vehicles currently have a cycle retention of at least 200-300 cycles of 80% of their capacity and a specific energy of 408 mA h kg^-1 (Sawicki and Shaw). An SIB with a 210-cycle life and 299-284 W h kg^{-1} specific energy has been reported (Sawicki and Shaw). Therefore, SIBs are quickly nearing the capabilities of LIBs and have the potential for widespread commercial use. They are also promising replacements for grid-scale electrical energy storage, as SIBs are cheaper than LIBs due to sodium's natural abundance (Sawicki and Shaw). For electric vehicles, a Na cathode has been reported to have 30,000 cycles with 95% capacity retention, with cathode and anode specific capacities of 145 mA h g^-1 after 1200 cycles and 400 mA h g^-1 after 150 cycles respectively (Sawicki and Shaw). Overall, while further advancements are needed for SIBs to fully replace LIBs, their outlook and recent progress is very promising.

While SIBs have potential, several issues prevent commercialization. Almost all SSEs suffer from issues like poor electrochemical properties, low ionic conductivity, and interface instability. All of these issues limit the applications of SIBs and need to be further researched if SIBs are to become widespread. Further insight into strong SEI formation could assist in creating stable SIBs, specifically, developing a mechanically strong SEI layer that can suppress dendrite formation (Hou et al.; Lu et al.). Many SPEs have been developed for this purpose, but if they are strong mechanically, they typically are insufficient in other areas including ionic conductivity and stability. This is a common issue that occurs in the development of SEIs and electrolytes in general. An ideal SEI layer needs to be strong mechanically, uniform, and prevent side reactions. Finding a way to balance all of these qualities is the key to safe, efficient SIBs. Further research should be conducted into the ideal SEI layer, methods to suppress dendrites, and increased ionic conductivity.

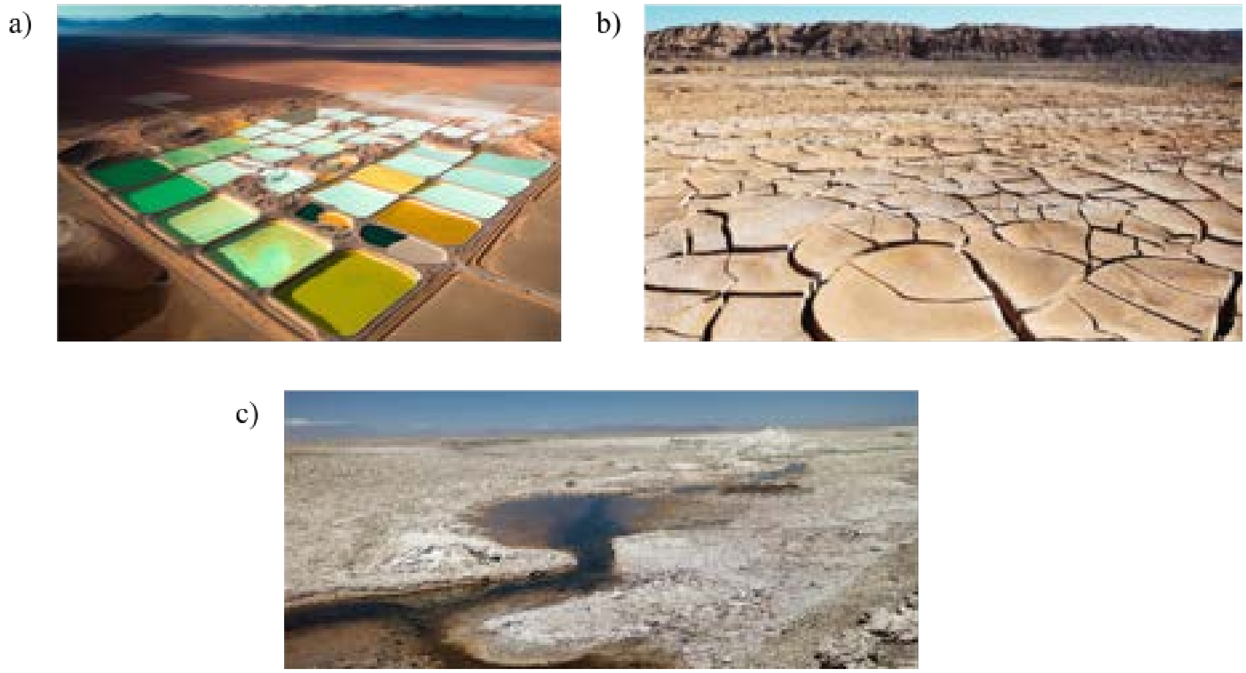

Fig 8: a) Picture of a lithium mine taken from Bloomberg (2018). b) Picture of Chile's Atacama region taken from Boddenberg (2018). c) Picture of environmental pollution from lithium mining taken from Boddenberg (2018).

LIBs have become an essential part of daily life in the past few decades. However, lithium is relatively scarce, so concerns that the natural lithium supply will run out have arisen. Moreover, lithium mining has detrimental consequences for the environment and locals. It releases greenhouse gasses, and lithium extraction uses chemicals that can leak into the soil and water (see Fig 6a), which can harm biodiversity in waterways (see Fig 6c). Moreover, the immense water usage can cause water shortages for the locals' consumption. In the Acatama region, the lithium mining is causing an already dry region to become dryer (see Fig 6b) (Boddenberg). Ana Ramos, president of the Council of Indigenous Peoples of Atacameños, said "the (lithium mining) company steals our water" (Boddenberg). In addition, cobalt, needed for $LiCoO_2$ batteries, is toxic to miners and cobalt mining partly takes place in politically unstable countries (Vignarooban et al.). Furthermore, lithium mining places heavy strain on the three countries (Argentina, Bolivia, and Chile) where lithium is primarily mined (Kaunda). Increased demand for lithium will require these countries to direct more resources toward lithium mining and less toward their people. On the other hand, sodium is naturally abundant and can be imported from many different countries, which would reduce the pressure put on Argentina, Bolivia, and Chile. A reduced need for lithium would mean less water would go to mining lithium and more to the citizens, improving their quality of life. Also, less toxic chemicals would pollute the soil, waterways, and air, allowing for a cleaner living environment. Overall, the living conditions of countries that are heavily reliant on lithium mining could improve from switching from LIBs to SIBs.

6. Conclusion

In this work, we reviewed past discoveries and recent developments on interface issues associated with sodium ion batteries (SIBs) with solid polymer electrolytes (SPEs) and gel polymer electrolytes (GPEs) and discussed how improvements could be made. SIBs are very promising as a cheap, environmentally friendly energy source. However, they suffer from interface issues that hinder their application. A deeper understanding of the dendrite formation, reactivity, and solid electrolyte interphase (SEI) formation can enable further development of SIBs. Overall, progress in developing electrolytes to overcome these obstacles is promising, but they mainly focus on only one issue. An ideal electrolyte needs to account for all limiting factors. Nevertheless, working toward effective and safe SIBs can improve environmental and humanitarian issues.

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